

### CLAIMS

We claim:

1. A method of producing one or more complex hydride compounds capable of reversible hydrogenation, comprising:

5           mechanically mixing a alkali metal hydride with aluminum powder and a powder of a transition metal catalyst compound in order to provide a compounded powder mixture; and

                  hydrogenating said compounded mixture at an elevated temperature and pressure to provide one of more alkali metal-aluminum hydride compounds.

10       2. The method according to claim 1, wherein said alkali metal hydride is selected from the group of hydrides consisting essentially of LiH, NaH, and KH.

3. The method according to claim 1, wherein said transition metal catalyst compound is selected from the group of compounds consisting essentially of  $\text{TiCl}_3$ ,  $\text{TiF}_3$ , and mixtures thereof.

15       4. The method according to claim 2, wherein the molar ratio of said alkali metal hydride to said aluminum powder is 1:1 to 4:1.

5. The method according to claim 3, wherein said molar ratio of the transition metal catalyst compound to the alkali metal hydride is 1:20 to about 1:100.

20       6. The method according to claim 1, wherein said step of mechanically mixing comprises a process selected from the group consisting of ball milling, plate or impact grinding, and blending, stirring, or agitating with or without a mechanical aid.

7. The method according to claim 6, wherein said step of mechanically mixing comprises ball milling said alkali hydride and said aluminum powders at a weight ratio of mill balls to said powders of 30:1 to 12:1 for a time of 0.1 to 10 hours.

**8.** The method according to claim **1**, wherein said step of mechanically mixing is carried out in an atmosphere consisting essentially of argon.

**9.** The method according to claim **1**, wherein said step of mechanically mixing is carried out at about room temperature.

5 **10.** The method according to claim **1**, wherein said step of hydrogenation is performed at an initial temperature of above about 60°C, and wherein said hydrogen pressure is maintained above an equilibrium plateau pressure for hydrogen at said initial temperature.

10 **11.** The method according to claim **1**, wherein said step of hydrogenation is performed at an initial temperature about 125°C, and wherein said hydrogen pressure is maintained at about 100 atmospheres and for at least about 2 hours.

**12.** A method of producing one or more complex hydride compounds capable of reversible hydrogenation, comprising:

15       mechanically mixing a comminuted form of an alkali metal, with aluminum powder and a powder of a transition metal catalyst compound to provide a compounded mixture; and

          hydrogenating said compound mixture at an elevated temperature and pressure to provide an alkali metal-aluminum hydride compound.

20 **13.** The method according to claim **12**, wherein said alkali metal is selected from the group consisting of Li, Na, and K.

**14.** The method according to claim **12**, wherein said transition metal catalyst compound is selected from the group of compounds consisting essentially of  $\text{TiCl}_3$ ,  $\text{TiF}_3$ , and mixtures thereof.

**15.** The method according to claim **13**, wherein the molar ratio of the alkali metal to the aluminum is 1:1 to 4:1.

**16.** The method according to claim **14**, wherein said molar ratio of the transition metal catalyst compound to the alkali metal is 1:6 to about 1:100.

5 **17.** The method according to claim **12**, wherein said step of mechanically mixing comprises a mechanical milling process selected from the group consisting of ball milling, plate or impact grinding, and blending, stirring, or agitating with or without a mechanical aid.

10 **18.** The method according to claim **17**, wherein said step of mechanically mixing comprises ball milling said comminuted alkali metal, said aluminum, and said transition metal catalyst compound at a weight ratio of mill balls to said mixed materials of 30:1 to 9:1 for a time of 0.5 to 3 hours.

**19.** The method according to claim **12**, wherein said step of mechanically mixing is carried out in an atmosphere consisting essentially of argon.

15 **20.** The method according to claim **12**, wherein said step of mechanically mixing is carried out at about room temperature.

**21.** The method according to claim **12**, wherein said step of hydrogenation is performed at an initial temperature above about 60°C, and wherein said hydrogen pressure is maintained above an equilibrium plateau pressure for hydrogen at said  
20 temperature.

**22.** The method according to claim **12**, wherein said step of hydrogenation is performed at an initial temperature of about 125°C, and wherein said hydrogen pressure is maintained at about 100 atmospheres for at least about 2 hours.

**23.** One or more complex alkali metal aluminum hydrides produced by the method of claim **1**, wherein said one or more complex alkali metal aluminum hydrides exhibit reversible hydrogenated and dehydrogenation states.

**24.** A hydride according to claim **23**, comprising  $\text{NaAlH}_4$  and  $\text{Na}_3\text{AlH}_6$ .

5 **25.** A method of providing a source of hydrogen gas comprising:

heating a quantity of an alkali metal aluminum hydride or hydrides produced by the method of claim **12**, to provide a supply of hydrogen gas and a dehydrogenated form of said alkali metal aluminum hydride; and

regenerating said alkali metal aluminum hydride by exposing said

10 dehydrogenated form of said alkali metal aluminum hydride to source of hydrogen gas and absorbing said hydrogen gas into said dehydrogenated form.

**26.** A method according to claim **25**, wherein said alkali metal is selected from the group consisting of Li, Na, and K.